MODELLING OF PARTICLE SURFACE MOISTURE CONTENT TO ESTIMATE GROWTH RATE IN FLUIDISED BED AGGLOMERATION

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KEYWORDS

Fluidisation, Mass transfer, Heat transfer, Simulation, Surface moisture, Agglomeration.

ABSTRACT

The objective of this study was to investigate the effect of particle surface moisture content on growth rate during agglomeration. In order to establish the relationship between process conditions and particle surface moisture content and temperature (and their spatial distribution in the fluidized bed), the combined population balance and thermodynamic model, as described by Ronsse et al. (2007a,b), was used. Growth kinetics were experimentally verified using 100 µm glass beads and maltodextrin based aqueous binder solutions, under varying process conditions. Studied process variables included binder concentration, fluidization air temperature, liquid binder feed rate and liquid binder atomisation pressure.

INTRODUCTION

Fluidised bed agglomeration is a widely used unit operation in the agrochemical, food and pharmaceutical industries with the aim of increasing particle size and consequently, to modify the behaviour of powders. In the fluidised bed agglomeration process, the binder, in the form of an aqueous or organic solution of a polymer, is continuously atomised onto the fluidised particles (Link & Schlünder, 1997; Zank *et al*., 2001). Particle growth is realised by means of interparticle liquid bridge formation (Link & Schlünder, 1997; Saleh *et al*., 1999). In order to solidify the interparticle liquid bridges, the bed is supplied with air having a high evaporative capacity (Depypere *et al*., 2003)

As the fluidised bed agglomeration process involves a series of complex thermodynamic interactions between the different phases involved, the agglomeration process is prone to yield-reducing or quality-degrading side-effects, i.e. uncontrollable particle growth and bed quenching (Guignon *et al*., 2002; Teunou & Poncelet, 2002). Although controlled agglomeration is the objective in many fluidised bed unit operations, in some applications agglomeration is also considered a negative side-effect (Dewettinck & Huyghebaert, 1998; Kage *et al*. 1998; Nakano & Yuasa 2001). For instance, in aqueous film coating, the preferred particle growth mechanism is layering, i.e. the binder is deposited on the particle surface without stable interparticle bridge formation. Whether growth by layering or growth by agglomeration is promoted, depends on process conditions including the fluidised bed's evaporative capacity (as a function of fluidisation air supplied to the bed) and the spraying rate of the dissolved binder, as is exemplified in Figure 1 (Gouin, 2005).

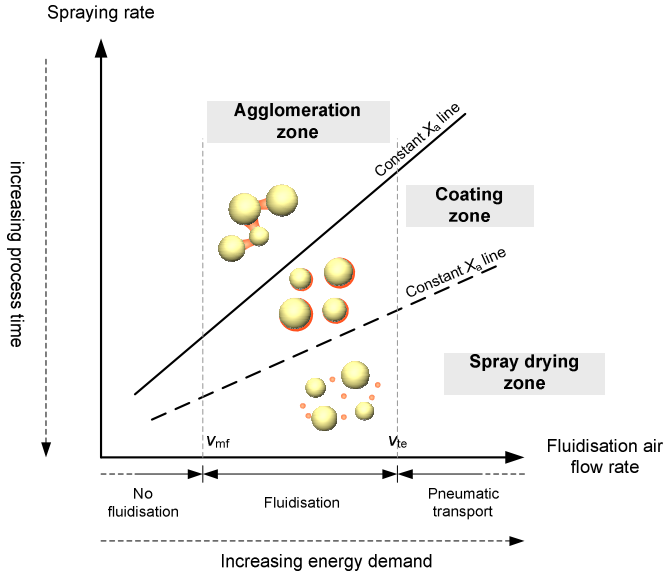

Figure 1. Relationship between particle growth kinetics in fluidised bed coating and the spraying rate and fluidisation air flow rate process variables (after Gouin, 2005).

One of the more common modelling methodologies for the fluidised bed agglomeration process is population balance modelling. The population balance equation is a statement of continuity that describes how the statistical distribution of one or more particle-related variables changes with time and space (Peglow *et al*., 2007). If the fraction of particles with size u at time t is given by the number density function $n(t, u)$, then the change in number of particles with size u as a result of binary aggregation of particles with size $(u - v)$ and v is given in the population balance equation as (Iveson, 2002; Peglow *et al*. 2007),

$$\frac{\partial n(t,u)}{\partial t} = \frac{1}{2}\int_0^v \beta(t,u-v,v)n(t,u-v)n(t,v)dv - \int_0^\infty \beta(t,u,v)n(t,u)n(t,v)dv \quad (1)$$

In the above equation is $\beta(t,u,v)$ the so-called *agglomeration kernel* and expresses the normalised frequency of collisions between granules with size u and v that result in binary aggregation. The problem with these kernels is that they are

one-dimensional and only relate particle size to aggregation probability.

With regards to fluidised bed agglomeration, it is clear that growth kinetics is not only influenced by particle size, but also by particle surface moisture content and to a lesser extent, particle temperature - as it, combined with surface moisture content influences the plasticity and viscosity of the binder on the particle surface. The objective of this study is the feasibility analysis of including particle surface moisture content and temperature into future agglomeration kernels.

MODEL DESCRIPTION

The model used in this study is a modified version of the fluidised bed coating model presented and validated by Ronsse *et al*. (2007a, b) and also used in scale-up studies by Hede *et al*. (2009). The model is based on the horizontal discretisation of the bed into *n* control volumes (or layers, S_i), each having a constant volume and containing a constant number of particles. Distinction is made between coating and non-coating control volumes. The atomised droplets have only a limited penetration depth in the fluidised bed, hence the model's definition of coating control volumes, which are the control volumes where droplet adhesion, mass (drying) and heat transfer occur simultaneously. In non-coating control volumes only mass (drying) and heat transfer occurs. A schematic overview of the model is given in Figure 2 – for more details, the reader is referred to Ronsse *et al*. (2007a).

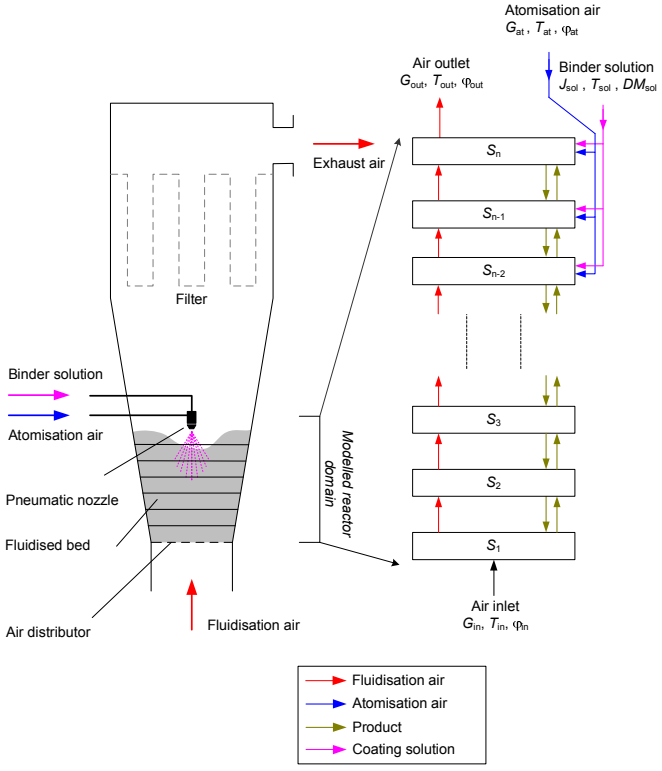

Figure 2. Scheme of the batch top-spray fluidised bed coater and the overall model, including mass flows of the gas phase (air), the solid phase (particles) and the sprayed coating solution.

Although the original model was intended for modelling coating processes, this model was selected for this study based on its possibility to predict the distributions of particle surface moisture content, total binder content and particle temperature.

The original model was modified so the binder solution dispersion on the particles momentarily residing in the coating control volumes was not continuous. Instead, the binder solution was dispersed in discrete quantities corresponding to a pre-set droplet volume among randomly selected particles, momentarily residing in the coating control volumes. Furthermore, the degree-of-wetting of the particles was also added to the model. The degree-of-wetting (ϕ_p) of a particle is defined as the ratio of wetted surface area (A_{wet}) to total particle surface area (A_p),

$$\phi_p = \frac{A_{wet}}{A_p} \tag{2}$$

In the model, for each timestep a number of particles in the coating control volumes are randomly selected for droplet-particle adhesion. If particle-droplet adhesion occurs, then the wetted particle surface area (A_{wet}) is increased by the droplet maximum spreading surface area,

$$A_{wet}(t+\Delta t) = A_{wet}(t) + \frac{\pi d_{\max}^2}{4} \tag{3}$$

In Equation (3) is d_{max} the maximum droplet spread diameter (m) after impacting on the particle substrate. The maximum droplet spread diameter was calculated using the Pasandideh-Fard *et al*. (1996) correlation,

$$d_{\max} = d\sqrt{\frac{\mathrm{We}+12}{3(1-\cos\theta_a)+4\left(\mathrm{We}/\sqrt{\mathrm{Re}}\right)}} \tag{4}$$

In equation (4) is *d*, the diameter of the droplet before impact (m), θ_a is the droplet/particle contact angle, *Re* is the droplet's Reynolds number and *We* is the droplet's Weber number, calculated according to

$$\mathrm{We} = \frac{\rho v^2 d}{\sigma},\quad \mathrm{Re} = \frac{\rho v d}{\mu} \tag{5}$$

Where ρ is the droplet's density (kg/m³), σ the surface tension (N/m), μ the viscosity (Pa.s) and *v* is the droplet impact velocity. In the model used in this study, the droplet impact velocity was assumed to be equal to the critical impingement velocity, v_{crit}. The critical impingement velocity is the velocity above which droplets no longer adhere onto the contacting substrate (i.e. the fluidised particles), it was calculated according to Link & Schlünder (1997),

$$v_{crit} = \frac{4\mu\left(3\tan(\theta_a/2)+\tan^3(\theta_a/2)\right)^{2/3}}{d\rho\tan^2(\theta_a/2)} \tag{6}$$

One of the limitations of the use of the degree-of-wetness is that once a particle is completely covered with liquid binder (i.e. $A_p = A_{wet}$), the degree-of-wetness no longer keeps track of the total liquid binder and surface moisture content (i.e. $\phi_p = 1$). Therefore, the particle surface moisture content was introduced as an absolute variable (W_p in mg/m²) along the degree-of-wetness. Similar to Equation (3), in case of droplet-particle adhesion, the particle surface moisture content is calculated as,

$$W_p(t+\Delta t) = W_p(t) + \frac{\pi d^3}{6}\rho(1-C) \qquad (7)$$

In Equation (7) is ρ the droplet's density (kg/m³) and C, the binder concentration of the droplet (kg/kg).

EXPERIMENTAL SET-UP

The experiments were performed in the top-spray reactor of the Glatt GPCG-1 fluidised bed unit (Glatt GmbH, Binzen, Germany), of which an overview is presented in Figure 3.

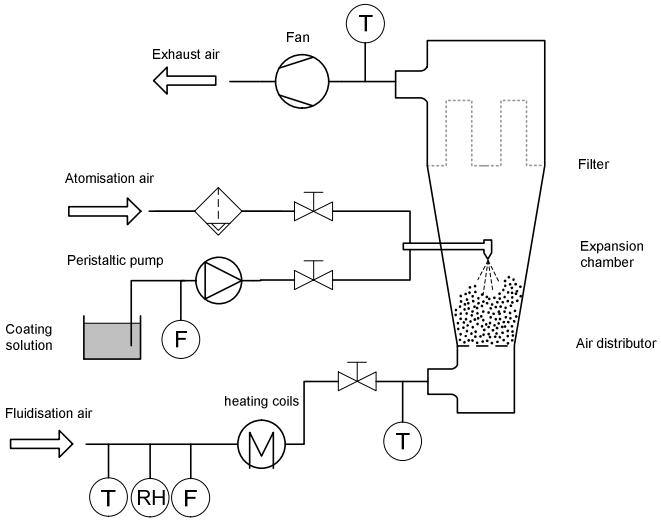

Figure 3. Schematic overview of the Glatt GPCG-1 fluidised bed coating unit.

Glass beads (Sovitec, Belgium) with a volume weighted average diameter of 110 µm and a density of 2650 kg/m³ were used as the core material. The binder consisted of a 10 w% aqueous solution of a hydrolysed starch, more specifically Maldex 18 (Tate & Lyle, Belgium). The glass beads were fluidised with heated air (75 °C) and with flow rates between 48.3 and 68.4 m³/h. Compressed air at 2.5 bar was used to atomise the binder solution.

In each batch experiment, 0.75 kg of glass beads were used and the process was terminated when the binder-to-core mass ratio reached 8 w% (i.e. 0.06 kg binder per 0.75 kg core material). Throughout the process (every 10 minutes) and upon completion, samples of the agglomerated core material were taken and particles size distribution was measured using laser diffraction analysis on a Malvern Mastersizer S equipped with a Qspec dry powder feeder. Based on the particle samples taken throughout the agglomeration process, the growth rate, G (in µm/min), was determined. An example of growth rate calculation for a single agglomeration process is illustrated in Figure 4. In this example, 7 particle samples were taken during the agglomeration process (on average every 10 min), and the volume-weighted average particle diameter, d_{43}, was determined using laser diffraction analysis. As can be seen from Figure 4, particle growth was almost linear (R^2 = 0.97 in the example) and therefore, growth rate (G in µm/min) was defined as the slope of the regression line in the d_{43} versus time plot. In the illustrated example, the growth rate was equal to G = 0.52 µm/min.

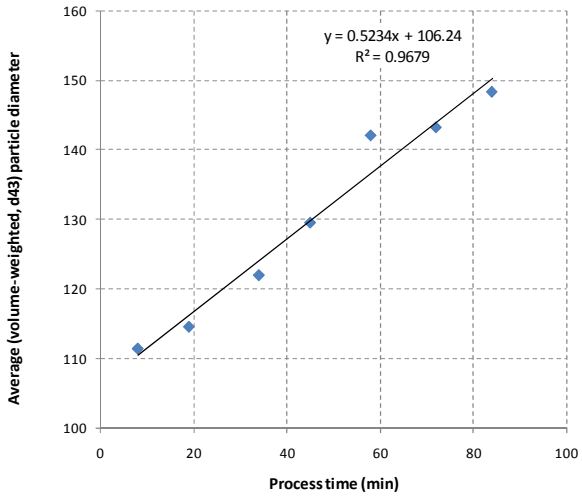

Figure 4. Example calculation of the growth rate, G (in µm/min) of an individual agglomeration experiment.

RESULTS AND DISCUSSION

In the agglomeration experiments, different series of experiments were performed in which the following parameters were changed: inlet air temperature, binder concentration, atomisation air pressure and binder spraying rate. Initial fluidisation air flow rates were kept constant for all experiments. Using the boundary conditions of each experiment, the process was simulated using the aforementioned model. Prior to analysing the growth kinetics, the overall model was validated using experimentally measured initial steady-state outlet air humidity, as shown in Figure 5. From these results, it can be seen that the model proposed in this paper approaches relatively closely (R^2 = 0.7) the experimentally measured values.

Next, the model-predicted surface moisture content during initial steady-state was compared against the experimental growth rate, based on particle size measurements at regular time intervals throughout the process, and is shown in Figure 6. The results suggest (R^2 = 0.61) that particle growth kinetics are positively influenced by surface moisture content. This effect is due to the available binder liquid to form interparticle liquid bridges. Although surface moisture content (as predicted by the model) is indicative for the volume of available binder on the particle, the model does not take into account the particles' binder concentration which strongly relates to interparticle bridge strength and

consequently, to growth kinetics. This could explain the rather weak correlation in Figure 6. With regards to particle temperature, a less clear correlation could be found between particle temperature and growth kinetics.

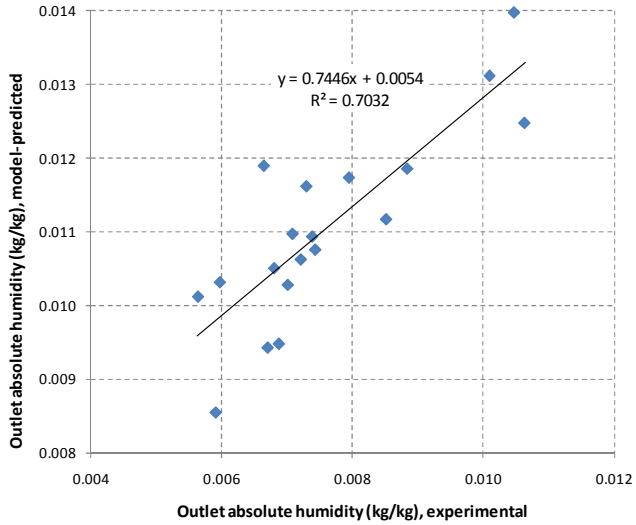

Figure 5. Modelled versus experimental outlet air absolute humidity (in kg/kg).

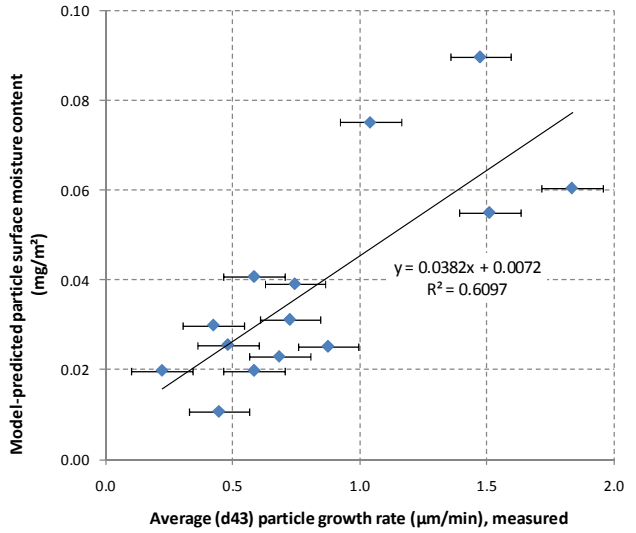

Figure 6. Model-predicted particle surface moisture content, W_p (in mg/m²) plotted versus experimental growth rate (in µm/min).

Although these results do suggest a moderate correlation between model-predicted surface moisture content and actual (experimental) growth rate, better understanding can be provided if the individual parameter variations are studied. In Figure 7, the effect of atomisation air pressure on the experimental growth rate and model-predicted surface moisture content is given. The model-predicted moisture content as a function of air pressure is not a smooth curve, due to some boundary conditions (like ambient temperature, inlet air relative humidity,…) not being constant in each modelled experiment. Higher atomisation air pressures result in a reduced particle growth rate due to smaller droplets being produced at higher atomisation air pressures. Smaller droplets evaporate more rapidly, reducing the amount of available liquid binder on the particle surface and hence, effectively reducing the growth rate. Considering atomisation air pressure, the model was able to predict the surface moisture content accurately and correlate it with the experimental growth rate.

Similar conclusions can be drawn from Figures 8 and 9, where the effects of liquid binder spraying rate (in g/min) and fluidisation air temperature (in °C) on the model-predicted surface moisture content and the experimental growth rate are plotted. In both cases, there is a strong correlation between the model-predicted surface moisture content and the actual (experimental) growth rate.

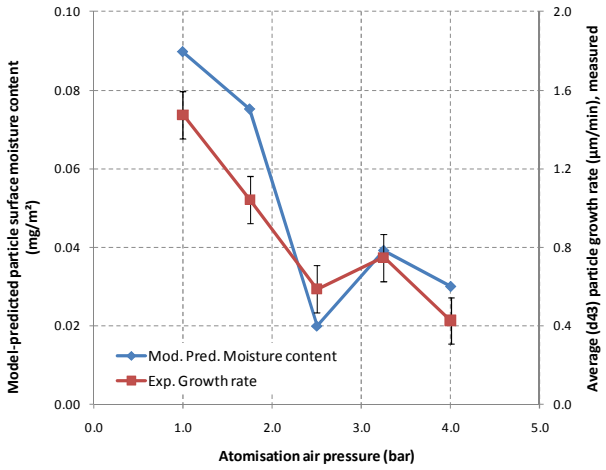

Figure 7. Model-predicted particle surface moisture content (mg/m²) and experimental growth rate (µm/min) versus atomisation air pressure (in bar).

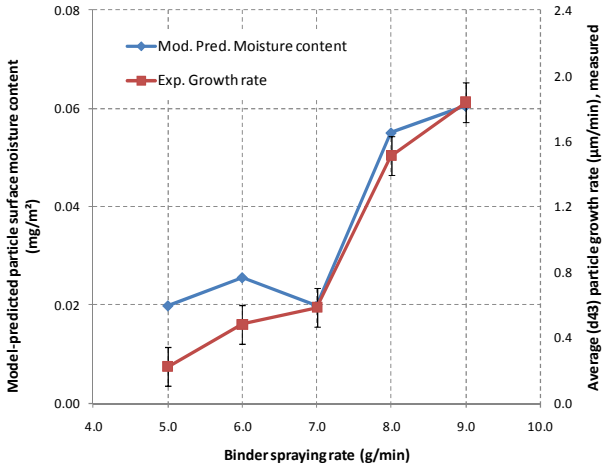

Figure 8. Model-predicted particle surface moisture content (mg/m²) and experimental growth rate (µm/min) versus liquid binder spraying rate (in g/min).

However, when studying the effect of binder concentration on the growth rate, as shown on Figure 10, the evolution of model-predicted surface moisture content is opposite to the experimental growth rate. Increasing the concentration of the binder will likely increase stickiness, or rather, the strength of the interparticle liquid bridges, formed during particle-particle collisions in the fluidised bed which will result in an

increase in growth rate. The model on the other hand, only accounts for the moisture in the binder which adheres onto the fluidised particles. As the concentration of binder increases, so decreases the moisture content, hence the reduction in predicted particle surface moisture content with increasing binder concentration.

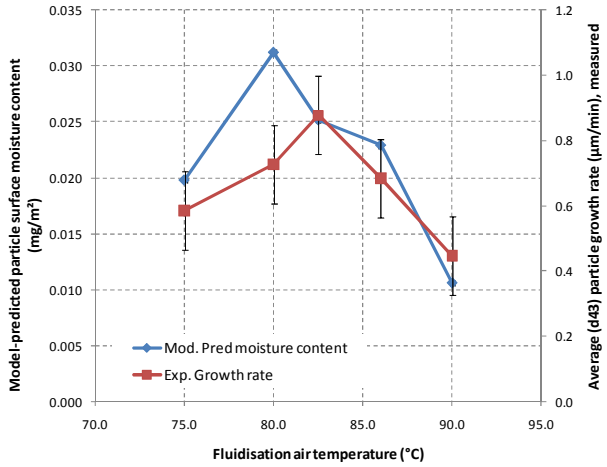

Figure 9. Model-predicted particle surface moisture content (mg/m²) and experimental growth rate (µm/min) versus fluidisation air temperature (in °C).

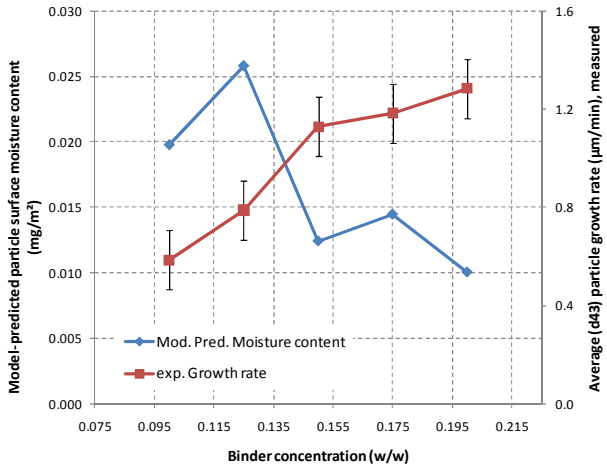

Figure 10. Model-predicted particle surface moisture content (mg/m²) and experimental growth rate (µm/min) versus liquid binder concentration (in kg/kg).

CONCLUSIONS

Based on these preliminary findings, it is clear that the particle surface moisture content is an important variable in modelling growth kinetics in fluidised bed agglomeration processes. A moderate to good correlation was found between model-predicted surface moisture content and actual growth rates. Therefore, in future population balance models for agglomeration, agglomeration kernels should take the modelled particle surface moisture content into account.

The importance of the particle surface moisture content with regards to particle growth could be explained by the required liquid binder on the particle's surface to form interparticle bridges during particle-particle collisions. However, the proposed model in this study does not account for the concentration of the dissolved binder of the freely available binder solution on the particle surface. The latter plays an important role in liquid bridge strength and hence, growth kinetics. Therefore, the proposed model is not yet capable of predicting agglomeration tendency with varying concentrations of liquid binder.

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BIOGRAPHY

FREDERIK RONSSE was born in Kortrijk, Belgium on November 3rd, 1978. He graduated from the Ghent University in 2001 with a M.Sc. in Biological Engineering Sciences. In 2006, he obtained his PhD degree in Applied Biological Sciences, on the development of heat and mass transfer models for fluidised bed coating processes in the food and pharmaceutical industries. His main research interests are multiphase flow analysis, including fluidisation, technology of granular matter and powders, study of heat and mass transfer, and modelling techniques including Computational Fluid Dynamics (PBM) and Population Balance Modelling (PBM) using Monte Carlo solving techniques.